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Imaging orbital-selective quasiparticles in the Hund's metal state of FeSe

Abstract

Strong electronic correlations, emerging from the parent Mott insulator phase, are key to copper-based high temperature superconductivity (HTS). By contrast, the parent phase of iron-based HTS is never a correlated insulator. But this distinction may be deceptive because Fe has five active d-orbitals while Cu has only one. In theory, such orbital multiplicity can generate a Hund's Metal state, in which alignment of the Fe spins suppresses inter-orbital fluctuations producing orbitally selective strong correlations. The spectral weights ?? of quasiparticles associated with different Fe orbitals ? should then be radically different. Here we use quasiparticle scattering interference resolved by orbital content to explore these predictions in FeSe. Signatures of strong, orbitally selective differences of quasiparticle ?? appear on all detectable bands over a wide energy range. Further, the quasiparticle interference amplitudes reveal that ??? < ??? ≪ ???, consistent with earlier orbital-selective Cooper pairing studies. Thus, orbital-selective strong correlations dominate the parent state of iron-based HTS in FeSe.

Disciplines

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Imaging Orbital-selective Quasiparticles in the Hund's Metal State of FeSe

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Strong electronic correlations, emerging from the parent Mott insulator phase, are key to copper-based high temperature superconductivity (HTS). By contrast, the parent phase of iron-based HTS is never a correlated insulator. But this distinction may be deceptive because Fe has five active d -orbitals while Cu has only one. In theory, such orbital multiplicity can generate a Hund's Metal state, in which alignment of the Fe spins suppresses inter-orbital fluctuations producing orbitally selective strong correlations. The spectral weights Z_m of quasiparticles associated with different Fe orbitals m should then be radically different. Here we use quasiparticle scattering interference resolved by orbital content to explore these predictions in FeSe. Signatures of strong, orbitally selective differences of quasiparticle Z_m appear on all detectable bands over a wide energy range. Further, the quasiparticle interference amplitudes reveal that $Z_{xy} < Z_{xz} \ll Z_{yz}$, consistent with earlier orbital-selective Cooper pairing studies. Thus, orbital-selective strong correlations dominate the parent state of iron-based HTS in FeSe.

The undoped phase proximate to superconductivity in copper-based materials is a strong Mott insulator^{1,2}, while that proximate to iron-based superconductivity is

generally of a metallic nature^{3,4}. This has motivated a perception that the mechanisms of high temperature superconductivity must be quite different in these two canonical materials classes and, moreover, that strong electronic correlations are not indispensable to HTS. Importantly, however, the electronic structure of the iron-based materials can still be governed by intense electronic correlations if an orbital-selective Hund's metal state exists⁵⁻¹³. This remarkable situation was discovered in theoretical studies of the multi-orbital Hubbard model (see the introduction to Supplementary Information) which typically consider the intra-orbital Hubbard energy U , the inter-orbital Coulomb interaction energy U' ($=U-2J$ for spin-rotational symmetry), and the inter-orbital Hund's interaction energy J between spins. For a range of strong J , dynamical mean field theory² (DMFT) predicts that inter-orbital charge fluctuations are greatly suppressed, leading to an orbital decoupling of the strong correlations⁶⁻¹³. The striking consequence is that strongly correlated and thus low coherence states associated with one orbital are predicted to coexist with coherent delocalized quasiparticle states associated with the other.

In theory, Hund's metals occur in a region of intermediate to strong U and of strong J ⁵⁻¹³. They are dominated by orbital-selective correlations, with the result that quasiparticle weights Z_m associated with different orbitals m diminish differently with increasing J or U . The quasiparticle weight Z is given by $Z(\mathbf{k}) = (1 - \partial \text{Re}\Sigma(\mathbf{k}, \omega) / \partial \omega |_{\omega=0})^{-1}$, where $\Sigma(\mathbf{k}, \omega) = \text{Re}\Sigma(\mathbf{k}, \omega) + i\text{Im}\Sigma(\mathbf{k}, \omega)$ is the self-energy of a quasiparticle state $|\mathbf{k}\rangle$ with momentum $\hbar\mathbf{k}$ that is subject to strong electron-electron interactions. The quasiparticle weight on band j in \mathbf{k} -space, $Z_j(\mathbf{k})$, can be connected to the quasiparticle weight in orbital space Z_m via the matrix elements of a unitary transformation. Multi-band Hubbard theories also exhibit orbital-selective quasiparticle (OSQP) phenomenology in which Z_m evolves differently for each orbital m ^{2,7,9-10,14-16}. Moreover, when orbital degeneracies are lifted, for example by crystal field splitting in an orthorhombic/nematic phase, this further suppresses inter-orbital charge fluctuations and amplifies the orbital decoupling that generates the OSQP^{7,9-11,14,17}. One approach to identifying such orbital-selective strong

correlations experimentally, would be to demonstrate that $Z(\mathbf{k})$ is highly distinct between the regions of the electronic bands that are associated with each different orbital m .

Because Fe-based materials supporting iron-based superconductivity are excellent candidates to exhibit Hund's metal orbital-selective effects, focus has naturally turned to detecting and understanding such phenomena in these systems. The resulting plethora of theoretical predictions^{6,8,11,13,15} include: (i) the electronic structure of iron-based superconductors should be heavily influenced by orbital-selective strong correlations, (ii) this effect is caused primarily by the Hund's decoupling of the interorbital charge fluctuations, (iii) the strength of correlations in each decoupled band $k_j(E)$ grows as it approaches half filling and, (iv) when orbital-selective strong correlations exist in such a state, Cooper pairing itself may become orbital-selective^{6,18-21}. Recent photoemission studies of orbital dependent bandwidth renormalization in these materials²² has been interpreted in this way. However a capability to directly visualize the orbital selectivity of the quasiparticles in the normal state of Fe-based HTS materials, ideally simultaneously with visualization of the electronic structures of the superconducting and nematic phases^{3,4}, remains an urgent priority.

To address this challenge, we focus on the compound FeSe, which shows clear indications of orbital selectivity^{6,15}. The FeSe crystal unit cell has $a=2.67$ Å, $b=2.655$ Å and $c=5.49$ Å in the orthorhombic/nematic phase below $T_S \cong 90$ K. Specifics of the Fe-plane of the same lattice can be described using the two inequivalent Fe-Fe distances $a_{Fe}=2.665$ Å and $b_{Fe}=2.655$ Å. The Fermi surface (FS) consists of three bands for which an accurate tight-binding model has been developed¹⁸. This model has excellent simultaneous consistency with angle resolved photoemission²³⁻²⁵, quantum oscillations²⁶⁻²⁸, and Bogoliubov quasiparticle interference^{18,19}. Surrounding the $\Gamma=(0,0)$ point is an ellipsoidal hole-like α -band, whose FS $\mathbf{k}_\alpha(E=0)$ has its major axis aligned to the orthorhombic b -axis; surrounding the $X=(\pi/a_{Fe},0)$

point is the electron-like ε -band whose “bowtie” FS $\mathbf{k}_\varepsilon(E = 0)$ has its major axis aligned to the orthorhombic a -axis; surrounding the $Y=(0,\pi/b_{\text{Fe}})$ point, a δ -band FS should also exist but has proven difficult to detect by spectroscopic techniques. Moreover, it was recently realized that orbital-selective Cooper pairing^{18,21} of predominantly the d_{yz} electrons causes the highly unusual superconducting energy gaps $\Delta_\alpha(\vec{k})$ and $\Delta_\varepsilon(\vec{k})$ of FeSe^{18,19}, from whose structure the FeSe quasiparticle weights are estimated to be $Z_{xy} \sim 0.1$; $Z_{xz} \sim 0.2$; $Z_{yz} \sim 0.8$ (with the other Z values being irrelevant for energies near $E=0$; Ref. 18). The challenge is to discover if all these exotic phenomena are indeed caused by the existence of orbital-selective strong correlations in a Hund’s metal normal state of FeSe.

Imaging of quasiparticle scattering interference²⁹ is an attractive approach. QPI has become widely used to determine exotic electronic structure of correlated electronic materials³⁰⁻³⁵. This effect occurs when an impurity atom/vacancy scatters quasiparticles which then interfere quantum-mechanically to produce characteristic modulations of the density-of-states $\delta N(\mathbf{r}, E)$ surrounding each impurity site; the global effects of this random impurity scattering are usually studied by using $\delta N(\mathbf{q}, E)$, the Fourier transform of $\delta N(\mathbf{r}, E)$. In a multi-orbital context, this can be predicted using

$$\delta N(\mathbf{q}, E) = -1/\pi \text{Tr} \left(\text{Im} \sum_{\mathbf{k}} \hat{G}_{\mathbf{k}}(E) \hat{T}(E) \hat{G}_{\mathbf{k}+\mathbf{q}}(E) \right), \quad (1)$$

where $G_{nm\mathbf{k}} = \sqrt{Z_n Z_m} G_{nm\mathbf{k}}^0$ with $\hat{G}_{\mathbf{k}}^0(E) = \left((E + i\eta) \hat{I} - \hat{H}_{\mathbf{k}}^{tb} \right)^{-1}$ is the electron’s Green’s function in orbital space, Z_m is the quasiparticle weight of orbital m , and $\hat{T}(E)$ is a matrix representing all the possible scattering processes between states $|\mathbf{k}\rangle$ and $|\mathbf{k} + \mathbf{q}\rangle$ for an impurity with on-site potential. Atomic scale imaging of these interference patterns $\delta N(\mathbf{r}, E)$ is achieved using spatial mapping of differential tunneling conductance $dI/dV(\mathbf{r}, E) \equiv g(\mathbf{r}, E)$, and has developed into a high-precision technique for measurement of electronic band structure $\mathbf{k}_i(E)$ of strongly correlated electron fluids³¹⁻³⁴. QPI should be of unique utility in searching for both orbital-selective coherence and spectral weight because: (i) the existence of

quantum interference itself is a robust test of \mathbf{k} -space coherence and (ii) the amplitude of QPI signals is sensitive as the squares of quasiparticle weights (Eqn. 1). Our target is thus to achieve orbitally resolved QPI from which the relative Z_m values of the normal state quasiparticles can be estimated.

We pursue this objective in the iron-based superconducting compound FeSe. Fig. 1a is a schematic representation of the orbitally resolved band structure of FeSe at $k_z=0$ (Ref. 18). Surrounding the $\Gamma=(0,0)$ point, the evolution of $\mathbf{k}_\alpha(E)$ is hole-like with the band top near $E=+15\text{meV}$ and d_{yz} orbital character (green) maximum along the x -axis while d_{xz} orbital character (red) prevails along the y -axis. Centered on the $X=(\pi/a_{\text{Fe}},0)$ point, $\mathbf{k}_\varepsilon(E)$ exhibits electron-like evolution with two Dirac points near $E=-25\text{meV}$, and d_{yz} orbital character (green) dominant along the y -axis while d_{xy} orbital character (blue) prevails along the x -axis. A fully coherent δ -band at the $Y=(0,\pi/b_{\text{Fe}})$ point would then have d_{xz} orbital character (red) dominant along the x -axis and d_{xy} orbital character (blue) prevailing along the y -axis. Fig. 1c,g show the orbitally-resolved constant-energy-contours (CEC) $\mathbf{k}_\alpha(E = -10\text{ meV})$ and $\mathbf{k}_\varepsilon(E = +10\text{ meV})$ of the α - and ε -bands in Fig. 1a. Fig. 1e,i then show the expectations based on Eq. (1) for the intraband QPI intensity patterns $|\delta N_\alpha(\mathbf{q}, E = -10\text{ meV})|$ and $|\delta N_\varepsilon(\mathbf{q}, E = +10\text{ meV})|$ corresponding to these contours, if all $|\mathbf{k}\rangle$ states are equally and fully coherent. If, by contrast, orbital-selective quasiparticles exist in FeSe, QPI should be very different because the quasiparticle weights Z_m associated with the Fe d -orbitals could all be distinct. In that situation, one might expect to see phenomena exemplified schematically by Fig. 1b. Here, for didactic purposes, we have chosen $Z_{xy} < Z_{xz} \ll Z_{yz}$. This means that in $\mathbf{k}_\alpha(E)$ the quasiparticle weight of d_{yz} orbital character (green) along the x -axis dominates strongly over the quasiparticle weight of d_{xz} orbital character (translucent red) along the y -axis (Fig. 1d). Similarly, for $\mathbf{k}_\varepsilon(E)$ the quasiparticle weight of d_{yz} orbital character dominates strongly along the y -axis compared to the negligible quasiparticle weight of the d_{xy} orbital character (pale blue) along the x -axis (Fig. 1h). The δ -band exhibits feeble quasiparticle weight of d_{xz} orbital character along the x -axis and negligible d_{xy} quasiparticle weight along

the x -axis. Under these circumstances, the QPI patterns will obviously be very different because scattering between regions with $Z_m \ll 1$ will produce far weaker intensity modulations. Thus, Fig. 1f,j show the anticipated intraband QPI intensity patterns $|\delta N_\alpha(\mathbf{q}, E = -10 \text{ meV})|$ and $|\delta N_\varepsilon(\mathbf{q}, E = +10 \text{ meV})|$, when the $|\mathbf{k}\rangle$ states have quasiparticle weights $Z_{xy} < Z_{xz} \ll Z_{yz}$. These are obviously quite different than those expected of fully coherent CEC in Fig. 1e,i and for the obvious reason that weak QPI intensity is produced by the quasiparticles of d_{xz} orbital character and virtually none by those of d_{xy} orbital character (SM Section II).

For FeSe, quantitative comparison of the QPI signature $\delta N(\mathbf{q}, E)$ expected for fully coherent bands versus strong orbital selectivity of quasiparticles, can then be carried out by using the T-matrix formalism. Here, the fully coherent Greens function $\hat{G}_\mathbf{k}^0(E)$ representing each $|\mathbf{k}\rangle$ state (a 5 by 5 matrix retaining orbital content information) is computed directly from the parameters of the electron band structure (Fig. 1a). These $\hat{G}_\mathbf{k}^0(E)$ are then used to calculate $\delta N(\mathbf{q}, E)$ from Eqn. 1. A scattering matrix $\hat{T}(E) = V_{imp} \hat{I} (1 - V_{imp} \sum_\mathbf{k} \hat{G}_\mathbf{k}^0(E))^{-1}$ representing $Z_m = 1$ for all m and a δ -function scattering potential at the origin in real space, and only $|\mathbf{k}\rangle$ for which $k_z = 0$, are used (SM Section II). Additionally, we numerically calculate the Fourier transform amplitude of the Feenstra transform, $L(\mathbf{r}, E) = N(\mathbf{r}, E) / \int_0^E N(\mathbf{r}, E') dE'$, to compare directly to measured normalized conductance, $(dI/dV)/(I/V)$ (see below and SM Section II). The resulting $|L(\mathbf{q}, E)|$ for fully coherent FeSe $|\mathbf{k}\rangle$ states are shown in Fig. 2a-d (and in Supplement Movie M1). These $|L(\mathbf{q}, E)|$ comprise QPI of α -, ε -, and δ -band for low \mathbf{q} scattering events. They show all the salient QPI features of fully coherent bands. By contrast, the QPI signatures of an OSQP in FeSe are determined using Eqn. 1 but with $G_{nm}(\mathbf{k}, E) = \sqrt{Z_n} \sqrt{Z_m} G_{nm}^0(\mathbf{k}, E)$ where $Z_m \in (0.073, 0.94, 0.16, 0.85, 0.36)$ for $m \in (d_{xy}, d_{x^2-y^2}, d_{xz}, d_{yz}, d_{z^2})$ and V_{imp} same as before (SM Section II). (Although these specific values chosen were taken from Ref. 18, the data in this paper as well as the data in Ref. 18 are consistent with the orbitally selective ansatz within a range of Z

values that are all consistent with the inequality $Z_{xy} < Z_{xz} \ll Z_{yz}$.) Most relevant are the orbitally resolved quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$ with the other two orbitals having negligible spectral weight near $E=0$. The predicted $|L(\mathbf{q}, E)|$ for OSQP are shown in Fig. 2i-l (and in Supplement Movie M2). These $L(\mathbf{q}, E)$ are now dominated by QPI of both α - and ε -bands as scattering in the δ -band is strongly suppressed due to decoherence of the respective quasiparticles. For the OSQP scenario, the scattering intensity distribution is strikingly C_2 symmetric. As expected, the QPI is dominated by quasiparticles with d_{yz} orbital content which are oriented along the k_x -axis in the α -band for $E < 0$ while being concentrated along the k_y -axis in the ε -band for $E > 0$. This produces the marked rotation of the QPI pattern by 90-degrees just above the chemical potential, a remarkable effect characteristic of FeSe³⁴ whose origin has until now proven elusive. Clearly the QPI predictions for OSQP (Fig. 2i-l and movie M2) are vividly different than those expected of a fully coherent conventional band structure (Fig. 2a-d and movie M1).

Our experimental search for OSQP phenomena uses spectroscopic imaging scanning tunneling microscopy (SI-STM) to study FeSe. The samples are inserted into the SI-STM instrument and cleaved in cryogenic ultrahigh vacuum at $T < 20\text{K}$. To focus on the normal state of FeSe, measurements for the energy range -8.75 meV to $+8.75\text{ meV}$ are acquired at $10.0\text{K} > T_c$, while the rest of the measurements are acquired at 4.2K to reduce thermal smearing. We have checked that the observed QPI phenomena do not differ between 4.2K and 10.0K (see SM section VIII). Differential tunneling conductance $g(\mathbf{r}, E) \equiv dI/dV(\mathbf{r}, E = eV)$ measurements are carried out with atomic resolution and register, as a function of both location \mathbf{r} and electron energy E . Because of the tiny areas of FeSe bands in \mathbf{k} -space (Fig. 1a), intraband QPI wavevectors are limited $|\mathbf{q}(E)| < 0.25(\frac{2\pi}{a_{Fe}})$, so that high-precision $g(\mathbf{r}, E)$ imaging in very large fields of view (typically $50 \times 50\text{ nm}^2$) is required. The Fourier transform of $g(\mathbf{r}, E)$, $g(\mathbf{q}, E)$, can then be used to reveal wavevectors and intensities of dispersive modulations due to QPI. However, to avoid artifacts (SM Section III) images of $L(\mathbf{r}, E = eV) \equiv \left(\frac{g(\mathbf{r}, E)}{I(\mathbf{r}, E)}\right)V$ are more typically used, and these

faithfully portray relative intensity at different directions in \mathbf{q} -space³⁴. Thus, Figs 2e-h show the measured $|L(\mathbf{q}, E)|$, the Fourier transform amplitude of $L(\mathbf{r}, E)$, from FeSe samples where the only scattering defects in the FOV are at Fe sites (topograph of measurement FOV is shown in SI Section IV and $|L(\mathbf{q}, E)|$ is provided as Supplement Movie). All such QPI data rotate by 90-degrees when measurements of $|L(\mathbf{q}, E)|$ are made in the orthogonal orthorhombic domain (SM Section V). Comparison of the measured QPI in Figs 2e-h to predicted $|L(\mathbf{q}, E)|$ for fully coherent bands (Figs 2a-d) and for OSQP (Figs 2i-l) reveals that the latter are in far better agreement. The intensity pattern and energy dispersion of the \mathbf{q} -vectors of maximum scattering intensity in measured $|L(\mathbf{q}, E)|$ closely follow those shown in Figs 2i-l, including the strong unidirectionality, and the sudden rotation of dispersion direction as $E=0$ is crossed. This provides a direct signature of OSQP in the metallic state of FeSe.

To visualize the impact of orbital selectivity on the complete band structure more globally, one can compare the energy dispersions continuously by comparing computed $|L(q_x, E)|$ and $|L(q_y, E)|$ to measured $|L(q_x, E)|$ and $|L(q_y, E)|$ respectively. For this purpose, Fig. 3a shows the theoretical dispersion of QPI maxima for α -, ϵ -, and δ -band along both q_x and q_y resolved by orbital content using the same color code as elsewhere. Fig. 3b shows the energy dependence of the predicted intensity of intraband scattering interference, along the same two trajectories as in Fig. 3a for fully coherent quasiparticle weights in all three orbitals $Z_{xy} = Z_{yz} = Z_{xz} = 1$. Fig. 3c shows the measured intensity of intraband scattering interference along q_x and q_y . The correspondence of these data to predictions in Fig. 3b is quite poor. However, in Fig. 3d we show the predicted intensity of intraband scattering interference if FeSe exhibits orbital selective QPI. The same two E - q planes as in Fig. 3b,c are shown, but now the OSQP quasiparticle weights are $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$. The correspondence between experimental $|L(\mathbf{q}, E)|$ (Fig. 3c) and the QPI signature of OSQP (Fig. 3d) is good and is discernibly superior to that with Fig. 3b.

If the quasiparticle weights indeed obey the relation $Z_{yz} \gg Z_{xz} > Z_{xy}$, it begs the question of whether weak QPI can be observed on the δ -band for its sections dominated by d_{xz} orbital content. Such phenomena should be clearest at states $E > 10 \text{ meV}$ (because the QPI from the α -band has disappeared here) and should appear along q_x due to scattering interference between d_{xz} dominated quasiparticles connected by a double-headed arrow shown in Fig. 4a. As seen in Fig. 4b, the expected scattering of states on the δ pocket is significantly suppressed. The remaining panels of Fig. 4 demonstrate that there is indeed a dispersive signal along q_x at somewhat higher q than the significantly stronger scattering interference along q_y from the d_{yz} sections of the ε -band. Detailed analysis and comparison of these two electron-like dispersive signals to simulation allow the conclusion that even the d_{xz} orbital content quasiparticles with very low Z_{xz} are detectable, as expected, on the δ -band (SM Section VI).

Finally, to visualize approximately how the $Z_j(\mathbf{k})$ evolve with \mathbf{k} -space angle around the Fermi surfaces of the α - and ε -bands, we measure the magnitude of $L(\mathbf{q}, E)$ on the \mathbf{q} -space trajectory through the QPI data for both bands. Fig. 5 a,b, show the measured angular dependence of QPI intensity for intraband scattering within the α - and ε -bands. The assignment of the scattering intensity to the electron and hole bands can be made by observing the dispersion of the intensity as a function of energy. In both cases, we focus on the trajectory of $\mathbf{q} = 2\mathbf{k}$ intraband scattering as indicated by the white crosses at which a local maximum in QPI amplitude is detected; the data are shown in full detail versus energy in SM Section VII. The $L(\mathbf{q}, E)$ amplitude is determined by taking line cuts through the measured $|L(\mathbf{q}, E)|$ maps (Fig. 5a,b) for a sequence of angles at a specified energy. Each line cut was fit to a sum of a linear background and a Gaussian peak to determine QPI signal amplitude (SM Section VII). The analysis was carried out for a sequence of energies (-25 meV to -15 meV for the α -band and +15 meV to +25 meV for the ε -band in 1.25 meV steps), and then the mean of these amplitudes (black dots in Fig. 5c,d) was taken over the relevant energy range ; the error bars represent the standard deviation of

an amplitude as energy is varied. Fig. 5c shows the measured $L(\mathbf{q}, E)$ intensity of α -band intraband QPI versus the \mathbf{q} -space angles from Fig. 5a integrated over energy range $-25\text{meV} \leq E \leq -15\text{meV}$ where this band is clear and distinct. Comparison to the theoretically predicted $L(\mathbf{q}, E)$ intensity (blue dot-dash curve) versus \mathbf{k} -space angle for orbitally selective QPI with $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$ (Section VII), reveals good agreement. Similarly, comparison of the measured ε -band $L(\mathbf{q}, E)$ versus the \mathbf{q} -space angles from Fig. 5b to the predicted $L(\mathbf{q}, E)$ intensity (blue dot-dash curve Fig. 5d) for orbitally selective quasiparticles having the same $Z_{xy}:Z_{xz}:Z_{yz}$ ratios (SM Section VII), yields $Z_{xy} \sim 0$. Therefore, the measured $L(\mathbf{q}, E)$ amplitude of QPI data (Fig. 5) are strongly consistent with orbital selectivity in the Hund's' metal quasiparticles of FeSe for which $Z_{xy} < Z_{xz} \ll Z_{yz}$.

The measured Z_m phenomena in Fig. 2-5 reveal the strength of orbitally selective strong correlations in the normal metal state of FeSe. The data indicate that this metal has delocalized $|\mathbf{k}\rangle$ states of d_{yz} character with good coherence because $Z_{yz} \sim 1$, $|\mathbf{k}\rangle$ states of d_{xz} character that are significantly less coherent, and $|\mathbf{k}\rangle$ states of d_{xy} character with lowest relative coherence. Comparison of measured $|L(\mathbf{q}, E)|$ to the theoretical $|L(\mathbf{q}, E)|$ predictions for different ratios $Z_{xy}:Z_{xz}:Z_{yz}$ (Fig. 2), along with evaluation of the \mathbf{k} -angle dependence of the QPI intensity for both bands (Fig. 5) (SM Section VII) all indicate that $Z_{xy} < Z_{xz} \ll Z_{yz}$. Moreover, we find the ratio of quasiparticle weights $Z_{xy}:Z_{xz}:Z_{yz}$ producing good agreement between theoretical $|L(\mathbf{q}, E)|$ and the QPI data $|L(\mathbf{q}, E)|$ (Figs. 2, 3, 4) to be indistinguishable from those deduced independently from the energy gap structure caused by orbital-selective Cooper pairing^{18,19}. This provides strong support for the concept of orbital-selective quasiparticle identification and Z quantification using QPI. Of most significance is that these orbital-selective QPI data provide direct demonstration that the normal state from which the HTS emerges in FeSe is dominated by orbitally selective strong correlations. If true in general for the iron-based HTS materials, this would be of fundamental significance because strong electronic correlations would then play a

central role in both copper-based and iron-based high temperature superconductivity.

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Author Contributions: A.K., Y.X.C. and P.O.S. developed and carried out the experiments; A.E.B. and P.C. synthesized and characterized the samples; A.K., P.O.S., and A.Kr., developed and carried out analysis; A.Kr., B.M.A. and P.J.H. provided theoretical guidance; B.M.A , P.J.H. and J.C.S.D. supervised the project and wrote the paper with key contributions from A.K., Y.X.C., P.O.S., A.Kr, and P.J.H. The manuscript reflects the contributions and ideas of all authors.

Figure 1 Orbitally resolved quasiparticle scattering interference in FeSe

- a.** Schematic representation of orbitally resolved band structure of FeSe at $k_z=0$. For each fully coherent quasiparticle state $|\mathbf{k}\rangle$ in these bands green represents d_{yz} orbital content, red represents d_{xz} orbital content and blue represents d_{xy} orbital content. The two Dirac points on the band surrounding the X-point $(\pi/a_{\text{Fe}}, 0)$ occur near $E = -25 \text{ meV}$ while the top of the hole-like band surrounding the Γ -point $(0,0)$ is close to $E = +15 \text{ meV}$
- b.** Schematic representation of the same orbitally resolved band structure of FeSe at $k_z=0$ but now indicating the effects of different quasiparticle weight Z . Here green represents the virtually fully coherent d_{yz} orbital content, translucent red represents the reduced Z value of d_{xz} orbital content and pale blue represents d_{xy} orbital content where Z tends towards zero.
- c.** Orbital content of constant-energy-contours (CEC) at the Γ -point $(0,0)$ at -10 meV using same color code as **a**.
- d.** Orbital content of CEC at the Γ -point $(0,0)$ at -10 meV using same color code as **b**.
- e.** Anticipated $|\delta N_\alpha(\mathbf{q}, E = -10 \text{ meV})|$ QPI signature of intraband scattering interference within α -band surrounding the Γ -point, for quasiparticle weights $Z_{xy} = Z_{yz} = Z_{xz} = 1$. The $|\delta N(\mathbf{q}, E)| \ (\equiv |1/\pi \text{Tr} \text{Im} \sum_{\mathbf{k}} \hat{G}_{\mathbf{k}}(E) \hat{T}(E) \hat{G}_{\mathbf{k}+\mathbf{q}}(E)|)$ images in panels E,F, I and J are calculated using T matrix with weak impurity potential using the band structure model displayed in panels **a** and **b**. In the calculations, the \mathbf{k} sum was restricted to the appropriate region of the Brillouin zone to separately capture intraband scattering interference pattern for different pockets.
- f.** Anticipated $|\delta N_\alpha(\mathbf{q}, E = -10 \text{ meV})|$ QPI signature for α -band with orbital-selective quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.
- g.** Orbital content of CEC at the X-point $(\pi/a_{\text{Fe}}, 0)$ at $+10 \text{ meV}$ using same color code as **a**.

- h. Orbital content of CEC at the X-point ($\pi/a_{Fe}, 0$) at +10 meV using same color code as **b**.
- i. Anticipated $|\delta N_\varepsilon(\mathbf{q}, E = +10 \text{ meV})|$ QPI signature of intraband scattering interference within ε -band for quasiparticle weights $Z_{xy} = Z_{yz} = Z_{xz} = 1$.
- j. Anticipated $|\delta N_\varepsilon(\mathbf{q}, E = +10 \text{ meV})|$ QPI signature for ε -band with orbital-selective quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.

Figure 2 Visualizing orbital-selective quasiparticle interference

- a-d** Predicted energy-resolved $|L(\mathbf{q}, E)|$ QPI signature of intraband scattering interference in a fully coherent state for quasiparticle weights $Z_{xy} = Z_{yz} = Z_{xz} = 1$. The white crosses correspond to $\frac{3}{16} \left(\frac{2\pi}{a_{Fe}}, \frac{2\pi}{b_{Fe}} \right)$ points in the momentum space. $|L(\mathbf{q}, E)|$ is the amplitude of the Fourier transform of the normalized conductance ($\equiv \frac{dI}{dV} / \left(\frac{I}{V} \right)$) at wavevector \mathbf{q} and energy E .
- e-h** Measured $|L(\mathbf{q}, E)|$ of FeSe at the same energies as shown in **a-d** and **i-l**. For all these energies, the measurements agree much better with the orbital-selective quasiparticle (OSQP) scenario (**a-d**) than with the fully coherent QPI predictions (**i-l**).
- i-l** Predicted energy-resolved $|L(\mathbf{q}, E)|$ QPI signature of intraband scattering interference in a OSQP with quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.

Figure 3 Energy dependence of orbital-selective quasiparticle interference

- a.** Momentum space representation of intraband quasiparticle interference maxima resolved by orbital content. Two E-q (energy-wavevector) planes are shown, parallel to q_x and to q_y . Color code shows the orbital content.
- b.** Predicted intensity of intraband scattering interference for a fully coherent state. Two E-q planes are shown, parallel to q_x and to q_y . The quasiparticle weights are $Z_{xy} = Z_{yz} = Z_{xz} = 1$.
- c.** Measured intensity of intraband scattering interference in FeSe. Same two E-q planes as in **b** are shown. Correspondence of these data to predictions in **b**

is poor, whereas their correspondence to the orbital-selective QPI prediction in **d** is much better.

- d.** Predicted intensity of intraband scattering interference for orbital-selective quasiparticles (OSQP) in FeSe. Same two E-q planes as in **b** are shown. The OSQP quasiparticle weights here are $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.

Images in panels **b,c**, and **d** are generated from q_x and q_y line cuts of the corresponding calculated and measured $|L(\mathbf{q}, E)|$. These cuts are normalized to unity for each energy to enhance visibility of band dispersions.

Figure 4 Detecting orbital-selective quasiparticle interference from both ε - and δ -bands above E_F

- a.** Quasiparticle constant energy contours at $E = +15 \text{ meV}$ showing the ε - and δ -bands. The color code indicates whether the quasiparticles are dominated by d_{yz} (green), d_{xz} (red) or d_{xy} (blue) orbital character. Double-headed arrows show scattering vectors along q_x and q_y .
- b.** Measured $|L(\mathbf{q}, E = +15 \text{ meV})|$ image. The directions of q_x and q_y line cuts are shown as blue lines. The signals from ε and δ bands are marked by a black circle and a black square, respectively. Signal locations were determined from fits to the line cuts (SI section VI). The white crosses correspond to $\frac{3}{16} \left(\frac{2\pi}{a_{Fe}}, \frac{2\pi}{b_{Fe}} \right)$ points in q space. $|L(\mathbf{q}, E)|$ is the amplitude of the Fourier transform of the normalized conductance ($\equiv (\frac{dI}{dV})/(\frac{I}{V})$) at wavevector \mathbf{q} and energy E .
- c.** E- q_y line cut through the sequence of measured $|L(\mathbf{q}, E)|$ images. The line cuts were fit to Gaussian peaks, and the locations of the peaks and the corresponding widths are shown as black circles with black lines.
- d.** E- q_x line cut through the sequence of measured $|L(\mathbf{q}, E)|$ images. The line cuts were fit to Gaussian peaks, and the locations of the peaks and the corresponding widths are shown as black squares with black lines. Note that

the maximum intensity in **d** is 50% of the maximum intensity in **c** with respect to the color bars.

Fig. 5 Momentum-angle dependence of orbital-selective quasiparticle weight Z_m

- a.** Measured $|L(\mathbf{q}, E = -20 \text{ meV})|$ image from the α -band showing the trajectory of the angularly resolved line cuts in **c**. Small white crosses mark the extracted peak location of QPI intensity, see SI section VII. The large white crosses correspond to $\frac{3}{16} \left(\frac{2\pi}{a_{Fe}}, \frac{2\pi}{b_{Fe}} \right)$ points in momentum space. $|L(\mathbf{q}, E)|$ is the amplitude of the Fourier transform of the normalized conductance ($\equiv (dI/dV)/(I/V)$) at wavevector \mathbf{q} and energy E .
- b.** Measured $|L(\mathbf{q}, E = +20 \text{ meV})|$ image from the ε -band showing the trajectory of angularly resolved line cuts in **d**. Small white crosses mark the extracted peak location of QPI intensity, see SI section VII.
- c.** Measured mean $L(\mathbf{q}, E)$ amplitude versus angle for the α band. The amplitudes are extracted from measured $|L(\mathbf{q}, E)|$ images on trajectory shown by crosses in **a**, and averaged over -25 to -15 meV energy range with the error bar showing the standard deviation for the sequence of amplitudes at different energies. Blue symbols show the predicted values of $|L(\mathbf{q}, E = -20 \text{ meV})|$ in the orbital-selective quasiparticle scenario with quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.
- d.** Measured mean $L(\mathbf{q}, E)$ amplitude versus angle for the ε band. The amplitudes are extracted from measured $|L(\mathbf{q}, E)|$ images on trajectory shown by crosses in **b**, and averaged over 15 to 25 meV energy range with the error bar showing the standard deviation for the sequence of amplitudes at different energies. Blue symbols show the predicted values of $|L(\mathbf{q}, E = +20 \text{ meV})|$ in the orbital-selective quasiparticle scenario with quasiparticle weights $Z_{xy} \approx 0.1$; $Z_{xz} \approx 0.2$; $Z_{yz} \approx 0.8$.

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